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LASER DAMAGE STUDY OF THIN FILMS

FOURTH QUARTERLY REPORT

FOR THE PERIOD

1 JANUARY 1967 - 1 APRIL 1967

Laser Damage Study of Thin Films

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FOURTH QUARTERLY REPORT

For the Period: 1 January 1967 - 1 April 1967

Report Prepared by: S. Refermat and A.F. Turner

ABSTRACT

A concentrated effort was made, during the fourth quarterly period, to increase the laser damage threshold, E_t, of aluminum oxide films. This objective was approached empirically. Selected vacuum deposition parameters were varied and the effect on E_t was observed. E_t for $\lambda/4$ and $33\lambda/4$ films were increased by a factor of two over previously reported values. The $\lambda/2$ threshold was increased by a factor of six over the previous value. The $\lambda/4$ threshold spontaneously fell to half its original value two days after manufacture.

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1.0 Introduction

The primary contract objective is the measurement of ruby laser energy densities needed to damage commonly used optical materials in thin film form. Since this primary goal has been achieved, we directed our efforts towards the secondary contract objective, which is to increase the established values of thin film laser damage thresholds, E_t (joules/cm^2). During the fourth quarterly period, we improved the threshold of aluminum oxide (Al_2O_3) films by varying selected deposition parameters. This report describes the results of our efforts.

2.0 Experimental

2.1 Sample Preparation and testing:

The thin film samples were produced by vacuum evaporation. The deposition was monitored using an optical system in the reflectance mode. The 99.3% pure aluminum oxide used in all the samples reported was obtained from the Norton Company, Worcester, Massachusetts. The white, grannular (grain size 0.2 - 0.8mm) material was easily evaporated by the electron gun with 2 kilowatt power level.

The 2" x 2" x 3/32" glass substrates have an index of refraction at λ = 5890A of 1.52. The substrate cleaning procedures used are listed in Table I.

The laser damage threshold measurements were performed using a previously reported technique.[1]

2.2 Optical thickness and absorption measurements:

The wavelength at which a sample had a $\lambda/4$ optical thickness was the wavelength of minimum transmittance. The wavelength of maximum transmittance was the wavelength at which the sample had a $\lambda/2$ optical thickness. The transmittance measurements were made on the Cary 14 recording spectrophotometer. The $\lambda/4$ transmittance values were used to obtain the index of refraction of the films, using an established method.[2]

Absorption measurements were made at the $\lambda/2$ position. The transmittance of the film-substrate combination should equal that of the substrate alone at the $\lambda/2$ position assuming a film homogeneous in refractive index. But absorption reduces the transmittance of the film-substrate combination. Values of absorption as little as 0.5% can be measured using this method.

3.0 Results

3.1 General Observations:

The damaged areas observed in high threshold ($E_t > 30$ joules/cm^2), and low threshold ($E_t < 20$ joules/cm^2), films differ in appearance. A typical high threshold damage spot has little symmetry. Its periphery consists of sharp, jagged irregularities. The boundary between the areas of film removal and residual film are sharp and distinct. Also, near threshold, the damage spot "radius" decreases rapidly with decreasing energy density. Low threshold damage spots are generally circular in appearance. Near threshold, the core area of complete film removal is not accompanied by substrate damage. The central area is usually surrounded by an annular ring of partial film removal. The damage spot "radii" of low threshold films decrease more slowly with decreasing energy density than those of high threshold films.

The mechanical durability of selected $\lambda/4$ samples was investigated. Essentially, the test apparatus was an eraser under a constant one pound load. To measure the mechanical durability, the number of rubs needed to permanently mar the film were counted. The values generally ranged from 750-1200 rubs. The film with the highest rub resistance also had the highest threshold ($E_t = 40$ joules/cm^2). A sample with an amount of absorption that could be detected by visual observation (film was tan colored in transmission), was tested also. It had a rub resistance of only 4 rubs.

The measured index of refraction values range from 1.59 - 1.61 ±0.02, in the visible region of the spectrum (Table II). These values were determined from $\lambda/4$ samples produced with the following values of parameters:

Pressure; $P = 1.8 \times 10^{-4}$ torr

Substrate temperature during deposition; $T_s = 170^{o}F$

Rate of Deposition; D = 12A/second

Electron gun power = 2.0 kilowatts

Previously, aluminum oxide films had been evaporated in an oxygen atmosphere. Oxygen was bled into the vacuum chamber to keep the pressure constant at 1.8×10^{-4} torr during evaporation. We decided to substitute an air bleed for the oxygen and observe the effect on E_t. The values of the other parameters were held constant. The resulting samples were compared and no measurable difference in laser damage threshold or mechanical durability was found. No difference in clarity was detected visually. The values of pressure quoted in this report refer to the residual air pressure in the chamber, not the oxygen pressure.

3.2 Threshold as a function of pressure.

The variations of threshold with pressure for three substrate temperatures during deposition are illustrated in Figures 1 - 3. All the samples measured to obtain these figures were produced at a deposition rate of 12A/second.

For the films tested to obtain Figure 1, the conditions held constant during evaporation were:

Substrate temperature during evaporation: $T_s = 120^{o}F$

Electron gun power level = 2 kilowatts

Optical thickness = $\lambda/4$ at λ = 5500 ±500A

Because the $\lambda/2$ position was below the glass cut-off, any absorption in these amples was not able to be measured spectrophotometrically. However, no absorption was detected with a visual observation. The maximum uncertainty in E_t is ±1½ joules/cm^2.

The parameters during evaporation of the samples of Figure 2 were:

T_s = 170°F

Electron gun power level = 2 kilowatts, except for the sample deposited at pressure = 2.5 x 10^{-4} torr. That sample was evaporated with a power level of 2.5 kilowatts to keep D = 12A/second.

Optical thickness = $\lambda/4$ at λ = 6900 ±700A

This graph has a definite maximum of E_t = 40 joules/cm^2. The $\lambda/4$ sample with the highest E_t also had the highest mechanical durability, 1200 rubs. This sample had no measurable absorption. The film evaporated at a pressure = 2.5 x 10^{-4} torr had 2% absorption at λ = 3600A. Any absorption in the other samples was unable to be measured on the spectrophotometer. Visually no absorption was detected. The maximum uncertainty in E_t is ±1½ joules/cm^2.

The parameter values used during production of the samples tested to obtain Figure 3 were:

T_s = 350°F

Electron gun power level = 2 kilowatts

Optical thickness = $\lambda/4$ at λ = 5500 ±500A

E_t increases slowly as the pressure is increased. One sample did have, and one did not have measureable absorption. Absorption in the other eight samples was unable to be measured on the spectrophotometer. No absorption was detected by visual inspection. The largest uncertainty in E_t is ±2.5 joules/cm^2.

3.3 Threshold as a function of substrate temperature during deposition, T_s:

The samples tested to obtain Figure 4 were deposited with the following conditions constant:

Pressure = 1.8 x 10^{-4} torr

D = 12A/second

Electron gun power level = 2.0 kilowatts

Optical thickness = $\lambda/4$ at λ = 6900A ± 600A

Figure 4 has a definite maximum E_t = 40 joules/cm^2. Three of the seven samples tested had no measureable absorption. The other films had no visibly detectable absorption. The maximum uncertainty in E_t is ±1½ joules/cm^2.

3.4 Threshold as a function of Deposition Rate:

The parameters held constant during production of the samples used in the investigation of deposition rate were:

P = 1.8 x 10^{-4} torr

T_s = 170°F

Optical thickness = $\lambda/2$ at λ = 6900A ±100A

The effect of deposition rate on threshold is difficult to study, primarily because at certain rates of deposition (3, 2.4 A/second) the resulting films were either absorbing or had a graded index. The most striking result of the deposition rate investigation is the six times increase in threshold of two $\lambda/2$ samples. At D = 4 A/second, E_t = 42, 45 were obtained. These two samples had no measureable absorption at λ=6900A. The maximum uncertainty in E_t was ±1 joule/cm^2.

3.5 Substrate cleaning procedures and electron gun current and voltage at constant power:

The samples used in these investigations were produced under the following conditions:

$P = 3x\ 10^{-5}$ torr

$T_s = 350^oF$

D = 12 A/second

Electron gun power = 1.5 kilowatts

The values of threshold were not altered measurably (E_t = 10-12 joules/cm^2) as the substrate cleaning procedure was changed. The variation of electron gun current and voltage at constant power did not change the threshold values, (E_t = 10-12 joules/cm^2). The current, voltage and power values investigated were:

Current (amps)	Voltage (volts)	Power (KW)
280	5000	1.4
440	3400	1.5
480	3200	1.5

There was no visible absorption in any of these samples.

3.6 Deterioration of threshold with time:

The $\lambda/4$ sample yielding the highest threshold was produced under the following conditions:

$P = 1.8 \times 10^{-4}$ torr

$T_s = 170^{o}F$

D = 12 A/second

Electron gun power = 2.0 kilowatts

The film was irradiated one day after it was produced and E_t = 40 joules/cm.^{2} The sample was tested again two days after it was deposited and the threshold had decreased to 23 joules/cm^2.

4.0 Recommendations

The following procedure is suggested for future study of threshold versus selected deposition parameters.

1. Establish the degree of absorption and optical thickness of each sample.

2. Establish a family of threshold versus pressure curves at various substrate temperatures during deposition.

3. From the data collected in Step 2, use a bootstrap technique to zero in on the temperature - pressure combination yielding the highest threshold.

4. At the pressure-temperature values found in Step 3, study the effect on E_t of other parameters such as:

   deposition rate

   evaporation rate

   electron gun power level

   electron beam spot size

   glow discharge

   angle of incidence of evaporant on substrate

   electron gun current, voltage; constant power

   cleaning procedures

5. As the best values of the other parameters are determined, insure that the previously determined pressure - temperature combination still yields the highest threshold value.

5.0 Conslusion

The mechanism of laser damage in thin films is not well understood, nor are the above described effects on the damage threshold of variations in the vacuum coating parameters attendant upon the preparation of aluminum oxide films. An attempt is being made to formulate a model to explain them, and this will be reported upon later.

We thank Duane Waterman and Gibb Nettles for their valuable aid in producing and evaluating the thin film samples.

This report was written by S. Refermat and A.F. Turner.

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1. Turner, A.F., et. al., Office of Naval Research Contract Nonr 4717(00), Final Technical Report, pages 3 - 4, (1966).

2. Chang, L., 9th Quarterly Report to Fort Belvoir, Contract DA-44-009-AMC-124 (T) Mod. 2, (1965).

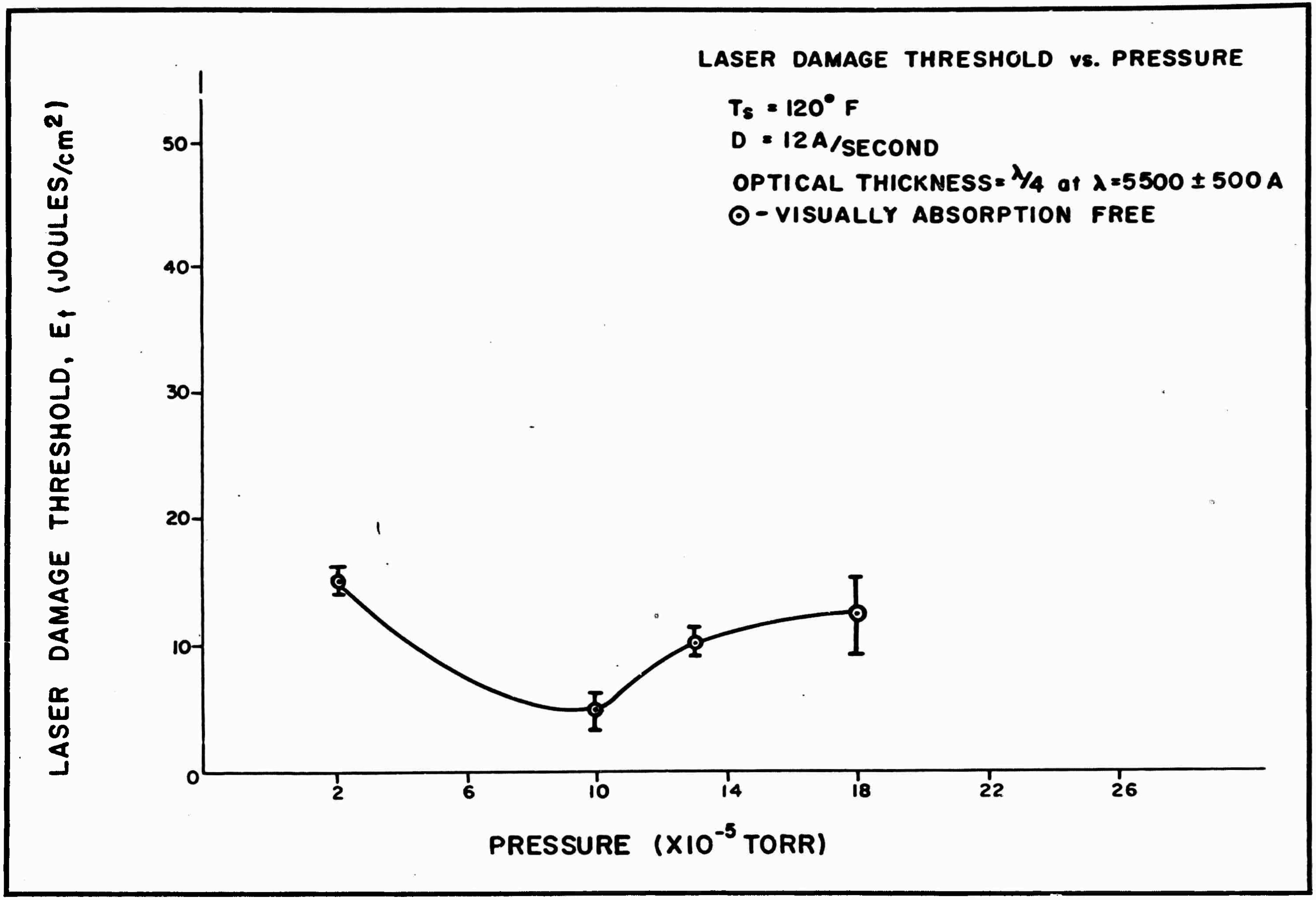

LASER DAMAGE THRESHOLD vs. PRESSURE
T_s = 120° F
D = 12 A/SECOND
OPTICAL THICKNESS = λ/4 at λ = 5500 ± 500 A
⊙ - VISUALLY ABSORPTION FREE
LASER DAMAGE THRESHOLD, E_t (JOULES/cm^2)
0
10
20
30
40
50
2
6
10
14
18
22
26
PRESSURE (X10^{-5} TORR)

FIG. 1

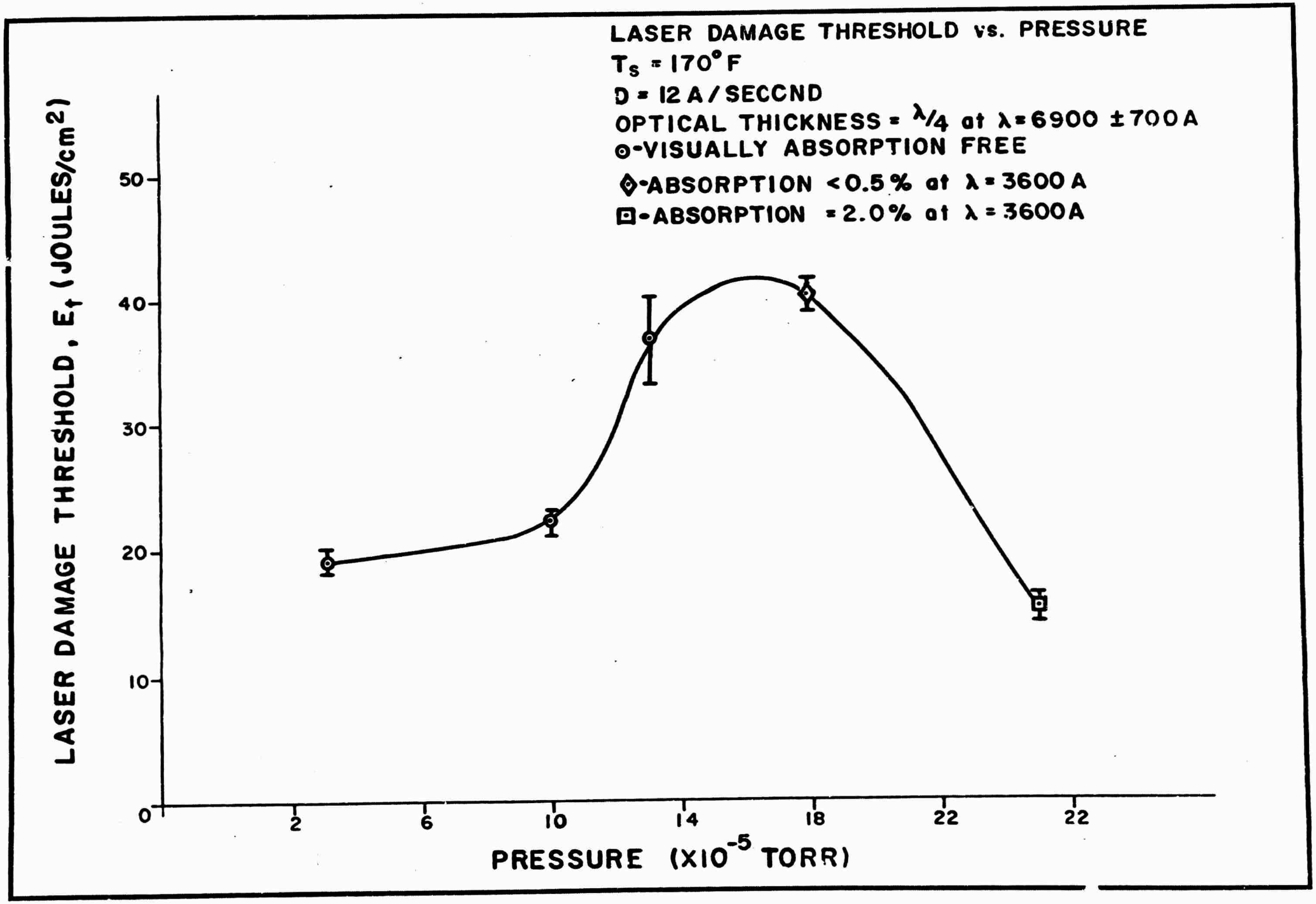
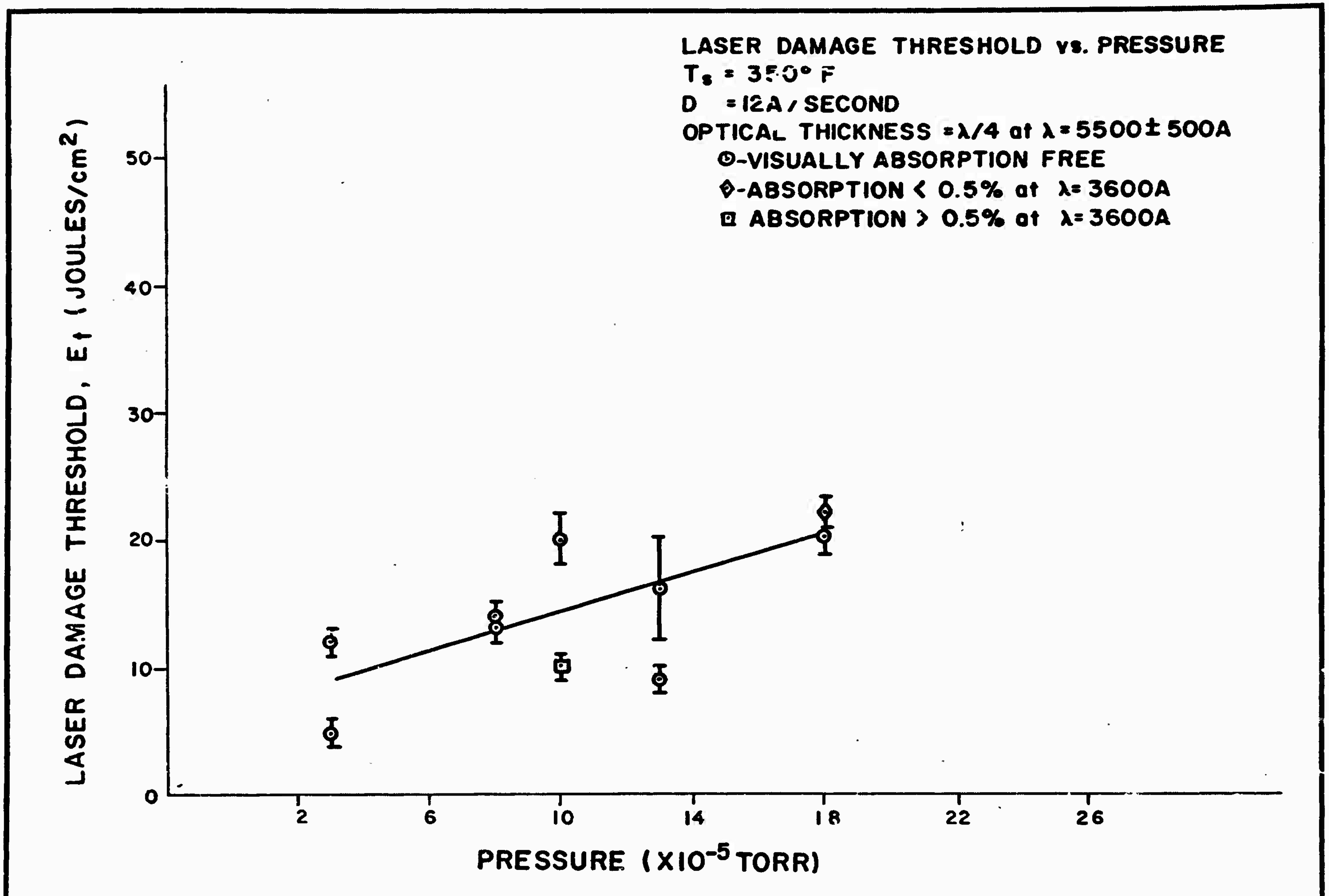

LASER DAMAGE THRESHOLD vs. PRESSURE
Ts = 170°F
D = 12 A / SECCND
OPTICAL THICKNESS = λ/4 at λ = 6900 ±700 A
⊙-VISUALLY ABSORPTION FREE
◇-ABSORPTION <0.5% at λ = 3600 A
⊡-ABSORPTION = 2.0% at λ = 3600A
LASER DAMAGE THRESHOLD, Et (JOULES/cm2)
50
40
30
20
10
0
2
6
10
14
18
22
22
PRESSURE (X10-5 TORR)

FIG. 2



LASER DAMAGE THRESHOLD vs. PRESSURE
Ts = 350° F
D = 12A / SECOND
OPTICAL THICKNESS = λ/4 at λ = 5500 ± 500A
⊙-VISUALLY ABSORPTION FREE
◇-ABSORPTION < 0.5% at λ = 3600A
⊡ ABSORPTION > 0.5% at λ = 3600A
LASER DAMAGE THRESHOLD, Et (JOULES/cm²)
0
10
20
30
40
50
2
6
10
14
18
22
26
PRESSURE (X10⁻⁵ TORR)

FIG. 3

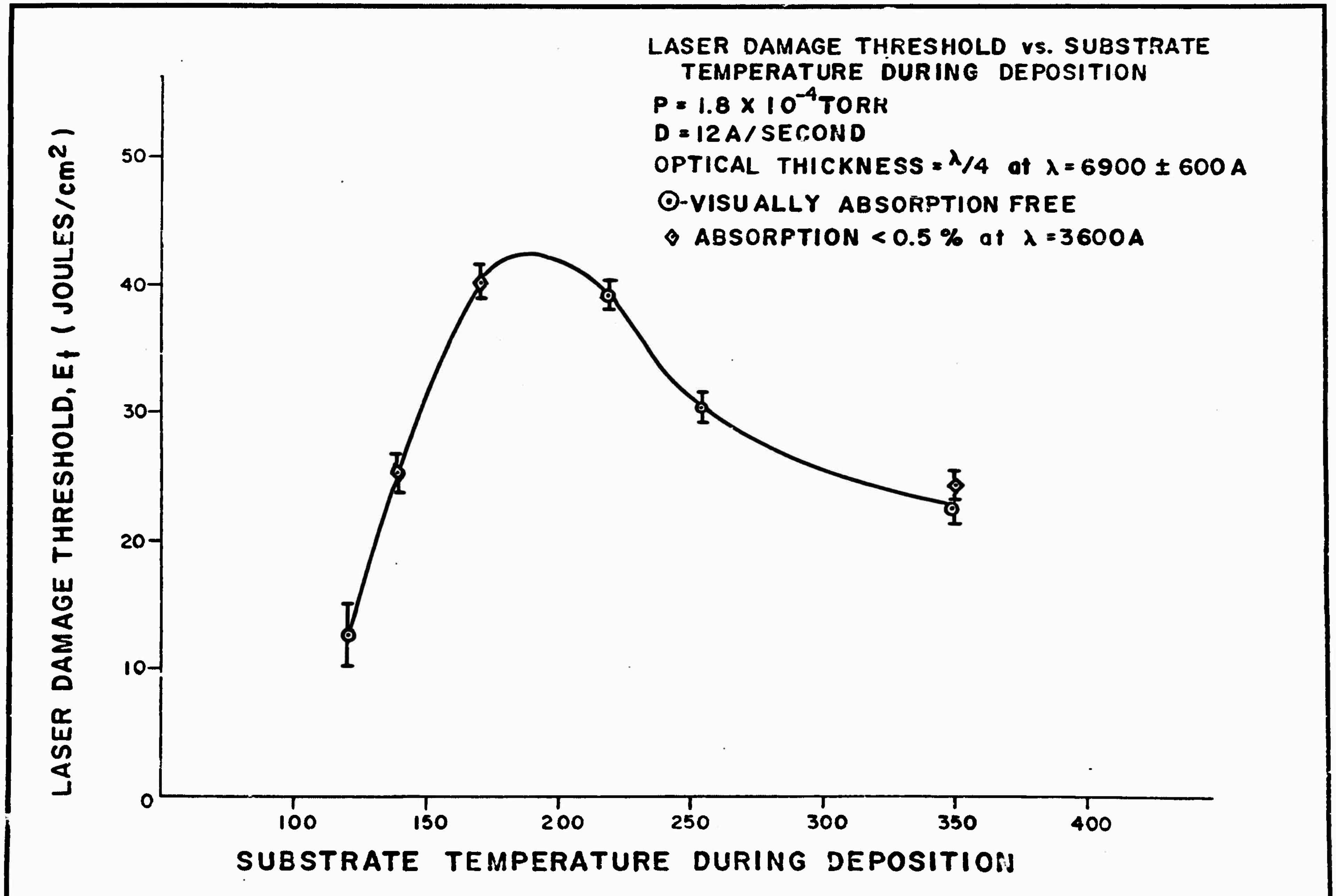

LASER DAMAGE THRESHOLD vs. SUBSTRATE TEMPERATURE DURING DEPOSITION
P = 1.8 X 10^-4 TORR
D = 12A/SECOND
OPTICAL THICKNESS = λ/4 at λ = 6900 ± 600 A
⊙-VISUALLY ABSORPTION FREE
◊ ABSORPTION < 0.5 % at λ = 3600A
LASER DAMAGE THRESHOLD, Et (JOULES/cm2)
0
10
20
30
40
50
100
150
200
250
300
350
400
SUBSTRATE TEMPERATURE DURING DEPOSITION

FIG. 4

TABLE I

COMPARISON OF SUBSTRATE CLEANING PROCEDURES

Ultra-Sonic Cleaning Method	Hand Cleaning Method
1. Ultra-sonic rinse in distilled water and detergent (orvious) at temperature = 150° F	1. Rinse in tap water.
2. Ultra-sonic rinse in distilled water at T = 150°F.	2. Polish with zirconium dioxide and wet cotton.
3. Rinse in tap water at 120°F	3. Rinse in tap water.
4. Rinse in alcohol	4. Dry with a soft, lint-free cloth
5. Rinse in the isopropyl alcohol degreaser at 180°F	

TABLE II

Index of Refraction versus
Wavelength*

Wave-length (μ)	Index†
0.44	1.59
0.55	1.61
0.69	1.60

*Samples used in index determinations were produced at:

Pressure = 1.8×10^{-4} torr

Substrate Temperature

During Deposition = 170°F

Deposition Rate = 12 Angstroms/second

Optical Thickness = $\lambda/4$ at wavelength indicated

†Index value uncertainty = ±0.02

TABLE III

Results of An Empirical Study to Increase E_t

CODE	SAMPLE	E_t (joules/cm^2)	OPTICAL THICKNESS
1	12/27/65 #2	26	$\lambda/4$ +
2	1/4/67 #1	22	
3	1/11/67 #1	40	
1	2/21/66 #34	7*	$\lambda/2$ +
2	2/8/67 #1	11	
3	2/15/67 #1	16	
4	3/22/67 #2	40-45	
2	1/4/67 #3	15	$6\lambda/4$ *
3	2/16/67 #2	12	$6\lambda/4$
1	3/2/66 #21	4.6	$36\lambda/4$ +
3	3/17/67 #1	9.0	$33\lambda/4$

*On silica substrate

+λ = 6900A

*λ = 5100A

CODE FOR TABLE III

1. Oxygen Pressure = 1.8×10^{-4} torr.

   Substrate Temperature
   During Deposition $T_s = 350^oF$

   Deposition Rate: D = 12 Angstroms/second

2. Residual Pressure: $P = 1.8 \times 10^{-4}$ torr.
   $T_s = 350^oF$

   D = 12 A /second

3. $P = 1.8 \times 10^{-4}$ torr.
   $T_s = 170^oF$

   D = 12A /second

4. $P = 1.8 \times 10^{-4}$ torr.

   $T_s = 170^oF$

   D = 4A /second

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13. ABSTRACT

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